

Self-organized Luminescent Nanostructures on Silicon Wafers

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Abstract- We report on the formation of luminescent nanostructures on the whole silicon wafer through a unique self-organization process. Nanoparticles of CdS form well-defined nanometer sized ring structures on the silicon wafer. Luminescence imaging shows one-to-one correspondance between topography and the light emission. These novel material systems are potential candidates for a variety of applications.

I. INTRODUCTION

Successful implementation of nanometer sized devices crucially depends on introducing a variety of functionalities to the nanostructures fabricated on the semiconducting wafers. There have been several techniques employed to address this problem. However, controlling the size, shape, functionality as well as the spatial distribution have been a problem. Recently, nanoparticles (NP) have been finding numerous applications in a wide variety of fields. They have been emerging as potential candidates for realizing controlled formation of functional nanostructures. We report here, a unique self-organization process of CdS NP, resulting in the formation of well-defined, luminescent nanometer sized ring structures (we call them, luminescent nanorings) on silicon wafer. CdS NP are introduced onto a silicon surface, pre-covered by a thin oxide layer, from an ethanol suspension. On annealing these samples in an ultrahigh vacuum chamber (UHVC), above the silicon oxide decomposition temperature ($>800^{\circ}\text{C}$), the

particles self-organize, leading to the formation of luminescent nanoring structures. Such nature driven processes are expected to play a vital role in the “bottom-up” approach, proposed to realize, futuristic nano devices. Further, the present work demonstrates the idea of incorporation of externally synthesized NP on to semiconductors (named as “plug and play”)^{1,2} to introduce functionality. We believe that this approach can make significant impact, as the particles can function as artificial atoms and/or quantum dots³ and has the potential to address the formidable challenges of realizing precise control of the size, shape, spatial location and the functionality of the nanostructures.

II. EXPERIMENTAL

Planar silicon wafers of (111) orientation (just, $\rho=5\ \Omega\text{cm}$) were used for the present experiment. Silicon wafers are cleaned thoroughly and a thin oxide layer ($<1\ \text{nm}$) is prepared by chemical means. NP are introduced onto this silicon wafer, covered with the thin oxide layer, at room temperature, by dip-coating¹ from ethanol suspension aided by ultrasonic bath. The samples are then loaded into the ultrahigh vacuum chamber and annealed to various temperatures, monitoring the nature of the surface species, *in situ*, by employing Auger, as well as photoelectron spectroscopic techniques. We have employed a variety of techniques such as small angle X-ray scattering, X-ray diffraction,⁴ scanning electron microscopy, Auger mapping, energy dispersive X-ray

analysis and extended X-ray absorption fine structure and atomic force microscopy (AFM) to characterize the system. Scanning tunneling microscopy (STM) measurements were performed independently in a separate instrument (Omicron). Optical characterization of the samples were carried out by photoluminescence (PL), scanning near field optical microscopy (SNOM) and tunneling electron luminescence (TL)⁵.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the pattern generated by the self-organization of CdS particles (10 nm) on the silicon surface after annealing. It consists mainly of nanometer sized ring structures. We also observe concentric rings formed across the surface steps and terraces. On the right hand side figure, we show a magnified version of the individual ring. The formation of the ring structures can be understood as a result of a drying or de-wetting process^{6,7} of the layer consisting of the CdS particles in it. Theoretical treatment of similar systems predict the formation of ripples which correspond to the concentric rings observed here⁸. Results of the STM examination are plotted in Figure 2 and indicate that the bare areas of silicon exhibit atomic resolution and the lattice fringes of CdS could be observed. These results indicate the well defined character of the system. As explained in reference 4, the size and shape of the particles could be controlled by controlling the parameters during the synthesis. As an example, we show the nature of the pattern formed from 25 nm sized particles, where we can form bigger rings filled with small size rings. Similarly, elongated cylindrical type particles join together on the surface during annealing resulting in the formation of star-like patterns as shown in Figure 3, indicating that the interparticle interaction also plays a role in deciding the nature of the pattern. Further, we can put physical constraints on the diffusion of these particles by selecting intentionally patterned silicon substrates as shown in Figure 4 (a). In this case, the particles are shown to organize around the edges of a mesa pattern.

Most importantly, we have mapped the luminescence from these ring structures by employing a powerful technique, called TL, where tunneling electrons are injected into the nanoparticles using a specially made conductive transparent tip which injects electrons into the nanoparticles and collects the luminescence signals simultaneously⁵ by a cooled photomultiplier tube and images by a tip-scan-synchronized photon counter. As shown in Figure 4 (b) and (c), emission from the particles self-organized in a ring structure is observed showing one-to-one correspondence between the morphology and the photon emission.

IV. CONCLUDING REMARKS

As mentioned in the introduction, the self-organization of these particles opens up the possibility of fabricating a whole variety of luminescent structures on silicon surface with possible applications in areas ranging from quantum optics to biotechnology^{9,10}. However, further work is required to fully understand this novel phenomenon which can lead to ultimate control of these nanostructures.

V. REFERENCES

- [1] K. Prabhakaran, K.V.P.M. Shafi, A. Ulman, T. Ogino, *Adv. Mater.* **2001** Vol. 13 No.24 1859-1861.
- [2] K. Prabhakaran et al. *Adv. Mat.* 2003 (In print).
- [3] P. Michler, A. Imamoglu, M.D. Mason, P.J. Carson, G. Strouse, S.K. Burotto, *Nature*. **2001** 406 968-970.
- [4] F. Meneau, S. Cristol, G. Sankar, I.P. Dolbnya, W. Bras, C.R.A. Catlow, J.M. Thomas, G.N. Greaves, *J. Appl. Cryst.* **2003** Volume 36, Part 3, p.718-721.
- [5] T. Murashita, K. Tateno, *Appl. Phys. Lett.* 78 (25) 3995 (2001).
- [6] M. Elbaum, S.G. Lipson, *Phys. Rev. Lett.* **1994** 72 3562.
- [7] E. Adachi et al., *Langmuir* 1995 11 1057.
- [8] A. Sharma and J. Mittal, *Phys. Rev. Lett.* **2002** 89 (18) p.186101.
- [9] M. Cai, O. Painter and K.J. Vahala, *Phys. Rev. Lett.* **2000** 85 (1) 74.

[10] C. Zandonella, *Nature* 423 p.10-12 (2003).

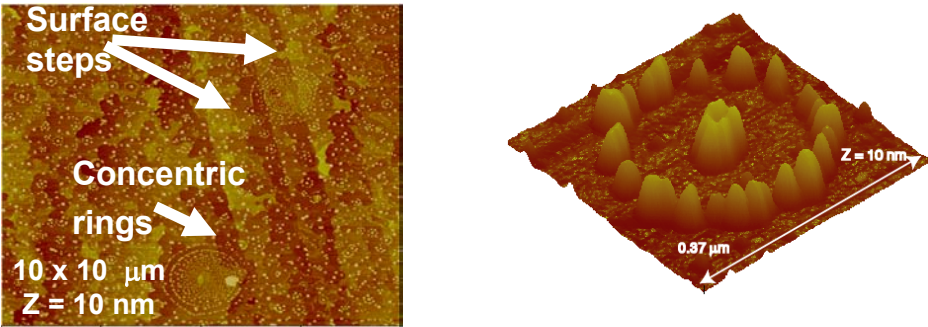


Figure 1 AFM image from self-organized CdS nanoring structures. An expanded version of a single ring is shown on the right side

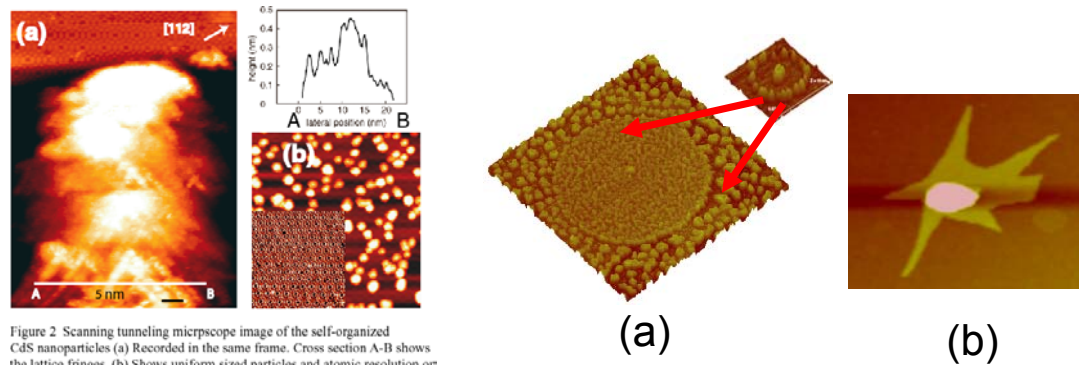


Figure 2 Scanning tunneling microscope image of the self-organized CdS nanoparticles (a) Recorded in the same frame. Cross section A-B shows the lattice fringes. (b) Shows uniform sized particles and atomic resolution on bare Silicon areas.

Figure 3 (a) AFM image from CdS (25 nm) particles on a silicon wafer (b) Unique pattern formed from elongated CdS particles.

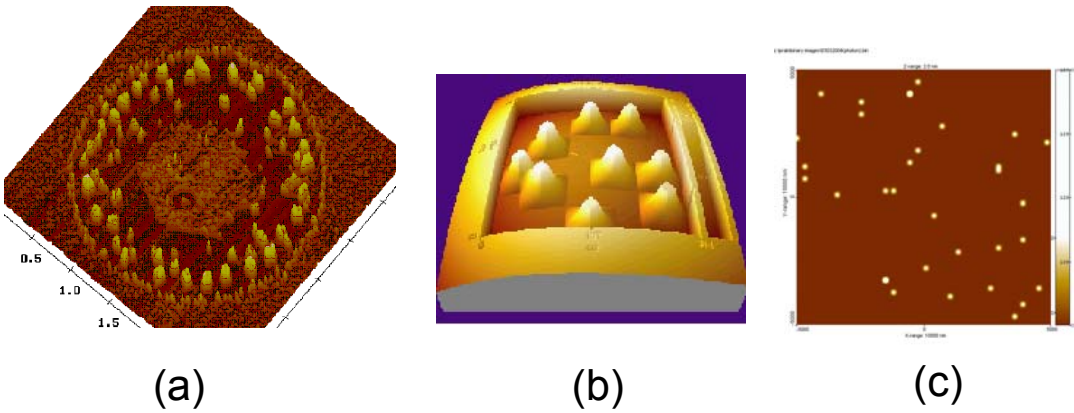


Figure 4 (a) CdS NP self-organized on a patterned substrate (image size 2 micron) (b) Tunneling luminescence images from the NP (b) and (c) 200 nm and 1 micron respectively.